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L1 ANSWER 1 OF 1 CA COPYRIGHT 2003 ACS

51:27873 Original Reference No. 51:5440g-i,5441a-b Stable dye baths. Hofer, Kurt; Hemmi, Hans M. (Sandoz Ltd.). CH 313159 19560515 (Unavailable). APPLICATION: CH.

AB Cations of the formula $\text{RCH}_2\text{CH}(\text{OH})\text{CH}_2(\text{N-A})_n\text{N}(\text{I})$, in which R is an alkoxy radical contg. at least 8 C atoms and may also contain oxygen, A is an alkylene residue with 2-4 C atoms, and n is an integer, are added to dye baths to reduce the speed of dyeing or redyeing. The cations are obtained by treating $\text{RCH}_2\text{CH}(\text{OH})\text{CH}_2\text{O}$, $\text{RCH}_2\text{CH}(\text{OH})\text{CH}_2\text{X}$, or $\text{RCH}_2\text{CHXCH}_2\text{OH}$ (where X = halogen) with polyamines of the formula $\text{HNR}'[\text{A-NR}'']_n\text{-1-ANR}'''\text{R}''''$, in which R', R'', R''', and R'''' are H, alkyl, aralkyl, hydroxyalkyl, or poly(glycol ether) radicals. Thus, $\text{C}_8\text{H}_{17}\text{OCH}_2\text{CH}(\text{OH})\text{CH}_2\text{O}$ (I) 1 mole is treated with $\text{H}_2\text{N}(\text{CH}_2\text{CH}_2\text{NH})_2\text{H}$ (II) 2 moles at 100-20.degree.. The excess II is distd. off in vacuo, the residue is treated with ethylene oxide (III) 8 moles at 100.degree., and subsequently at 50-60.degree. with Me_2SO_4 3 moles. The mixt. is heated 2 hrs. at 80-5.degree. and yields a brownish, viscous, H_2O -sol. product. Replacing the octyl radical in I with dodecyl, 9-octadecen-1-yl, isooctylphenyl, methylcyclohexyl, etc., varying the amts. of III or replacing III with $\text{ClCH}_2\text{CO}_2\text{H}$, or replacing II with $\text{H}_2\text{N}(\text{CH}_2\text{CH}_2\text{NH})_3$ or 4 H, 3-isopropylamino-1-propylamine, or 3-dimethylamino-1-propylamine yields similar products. A small addn. (2.5-12%) of these products to dye baths or vats insures level dyeing. A bath contg. 30% NaOH 15 cc., NaHSO_3 5 g., a l 0.45 g., and 9-octadecen-1-yl polyglycol ether (20 $\text{CH}_2\text{CH}_2\text{O}$ groups) 0.55 g. in H_2O 1 l. at 95-100.degree. removed within 30 min. the unevenly distributed dye on a 25 g. cotton-cretonne fabric.

L2 ANSWER 1 OF 1 CA COPYRIGHT 2003 ACS

87:151695 Bactericidal alkylene polyamines. Ito, Hirohiko; Mizuno, Yasushi; Yamamoto, Tsuneo (Takemoto Oil and Fat Co., Ltd., Japan). Jpn. Tokkyo Koho JP 52018047 B4 19770326 Showa, 5 pp. (Japanese). CODEN: JAXXAD. APPLICATION: JP 1972-114766 19721117.

AB $R_1R_2NZ(NR_3Z)_nNR_4R_5$ (I; $R_1-5 = H$, 3-alkoxy-2-hydroxypropyl; $Z = C_2-3$ alkylene, $n = 1, 2$), effective bactericides and fungicides against *Salmonella typhosa*, etc., were prep'd. by reaction of $H_2NZ(NHZ)_nNH_2$ (II) with alkyl glycidyl ethers. Thus, 29 parts II ($Z = CH_2CH_2$, $n = 1$) was heated with 70 parts 2-ethylhexyl glycidyl ether at 180.degree. to give the corresponding I, the HCl salt of which was 90 times more effective against *Salmonella typhosa* than was PhOH.

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(54) New Disinfectant Surfactant

"Bactericidal Surfactants"

(21) Application No. S42-35350

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(22) Application date: June 3, 1967

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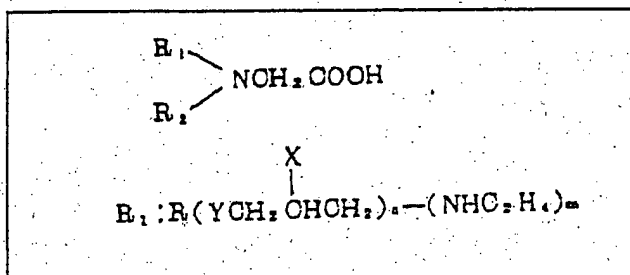
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Detailed Explanation of the Invention

This invention relates to a new disinfectant surfactant that has sterilizing power.

The disinfectant surfactant of this invention is generally comprised of the compound that can be indicated by the below listed general formula, and an inorganic or organic

hydrochloric acid (*this might be a typo of salt, Translator*) or an alkali salt of these.



R_1 :

R_2 : Hydrogen atom or R_1

[Here, R is an alkyl group with 6 to 16 carbon atoms, or an alkyl allyl group. Y is an oxygen atom or a sulfur atom. X is a hydroxyl group or a halogen atom. n is an integer of 1 to 3, and m is an integer of 1 to 5.]

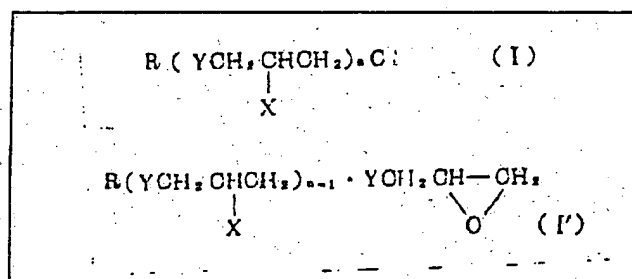
Until now, those that are known as the disinfectant surfactants are anionic phenol compounds, cationic quaternary ammonium compounds, and ampholytic alkyl polyalkylene poly amino acetic acids, etc. However, although these have quite good germicidal ability for the Gram-negative bacteria or the Gram-positive bacteria, the phenolic disinfectants have the side effects such as irritation of the skin, etc., and in addition, they do not have the cleansing power. On the other hand, the cationic disinfectants also do not have almost any cleansing power when they are used alone. It is not necessary to talk about anionic surfactants, but the blending of non-ionic surfactants is also not preferable, and also, skin irritation is severe too. Therefore, these are not suitable for the practical uses which perform disinfecting as well as cleansing. The above mentioned ampholytic surfactants can be blended with the non-ionic surfactants too, and the germicidal strength is excellent too, however, the side effects such as skin irritation, etc., can not be avoided, and that is a problem, and this is the present situation.

Considering the above mentioned present situation, the inventors of this invention studied for a long time to eliminate this problem, and they arrived at this invention as the result.

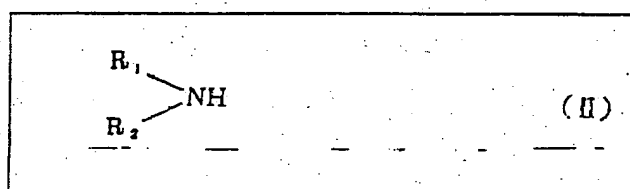
The above mentioned compound of this invention has the surfactant ability and it has strong germicidal ability for various pathogenic bacteria, micro organisms, and also for the non- pathogenic micro organisms in a wide range of hydrogen ion concentrations due to its unique molecular structure. In addition, these compounds have less toxicity, and they hardly cause any skin irritation, so that they have not only a wide range of usages, but also they can be mix- used with non- ionic agents since the co-use of it does not cause a reduction of the effect which is the case for the cationic disinfectants. Therefore, the non-ionic surfactants can be blended into the compound of this invention depending on necessity, and it can be utilized as an excellent sterilizing, disinfectant cleansing agent, and it can be applied very effectively for the environmental hygiene in a wide range of general sterilization of hands and fingers, washing and disinfecting of dishes, and washing and disinfecting the equipment and devices of the food industry, etc.

Next, the method to synthesize the disinfectant surfactant of this invention will be indicated. Namely, the compound (I) or compound (I') which can be indicated by the general formulas (I) and (I'), and which can be obtained by ordinary methods such as reacting a high rank alcohol or a high rank thio alcohol and epi chloro hydrin, etc., with a poly alkylene poly amine, and the compound (II) which can be indicated by the general formula (II) is obtained. And mono chloro acetic acid is reacted with the compound (II), and the compound (III) which can be indicated by the general formula (III) is obtained. Then the compound (III) is neutralized by an alkali, or the above mentioned compound (II) is reacted with a mono chloro acetic acid salt, and the compound (IV) is obtained. Then, the alkali is exposed to the above mentioned

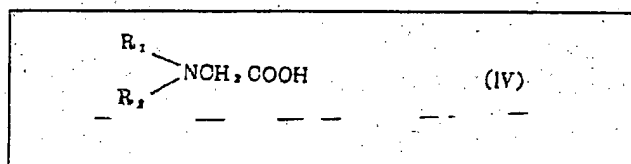
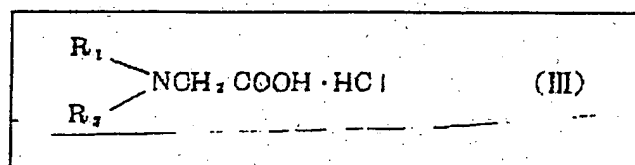
compound (IV), and the compound $(R_1)(R_2)NCH_2COOMe$ (both R_1 and R_2 are bonded to the N atom, Translator) can be obtained. [Here, Me is an alkali metal.]

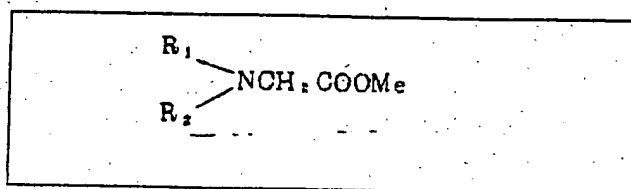


[Here, R, X and Y are the same as in the above description.]



[Here, R_1 and R_2 are the same as in the above description.]





Next, this invention will be explained referring to actual examples. In the actual examples, parts means weight parts.

Actual Example 1

111 parts of 2-octyl-2'-chloro hydrin ether was slowly dropped into 77 parts of diethylene tri amine at 160 °C. After the dropping was completed, it was reacted for 3 hours at 180 °C. After the reaction was completed, the contents were transferred into a separation funnel, and the amine hydrochloric acid salt that was by-produced was removed, and thus, the intermediate material which is mainly N, N'-bis (3-octa oxy -2-hydroxy propyl) di-ethylene tri amine, can be obtained.

9.5 parts of mono chloro acetic acid was dissolved in 50 parts of iso propanol, and this solution was slowly added into 0.1 mol of this intermediate material, and it was reacted for 2 hours at 80 to 90 °C, and N, N'-bis (3-octa oxy -2-hydroxy propyl) di-ethylene tri amino acetic acid hydrochloric acid salt (from now on, this will be called DOD -11) was obtained.

DOD - 11 was reacted with a sodium hydroxide solution, and the sodium chloride was removed, and the N, N'-bis (3-octa oxy -2-hydroxy propyl) di-ethylene tri amino acetic acid - sodium salt (from now on, this will be called DOD -13) was obtained.

DOD -11 (in the case when the octyl group is an - octyl group)

The surface tension (0.25 % solution, 20 °C stacto method) was 23.56 dyne / cm
foaming strength (0.25 % solution, 20 °C Losmiles? (*phonetically written*) method) 215 mm

Disinfectant strength (phenol coefficient) is indicated in Table 1.

Table 1. Disinfecting strength of DOD - 11 (in the case when the octyl group – octyl group.)

Composition of the material tested		Bacteria used	Phenol coefficient
DOD - 11	Non-ionic surfactant*		
10 %	0 %	Bacterium typhosum	38
10 %	3 %	Bacterium typhosum	39
10 %	3 %	Escherichia coli	33
10 %	3 %	Yellow Staphylococcus	7.1
10 %	3 %	Dysentery bacillus	50
10 %	20 %	Bacterium typhosum	39

* Non- ionic surfactant = poly oxy ethylene ($\bar{P} = 8$) octyl phenyl ether

DOD -11 (in the case when the octyl group is a 2- ethyl hexyl group)

The surface tension (0.25 % solution, 20 °C stacto method) was 26.69 dyne / cm

The phenol coefficient of a 10 % solution for typhoid bacteria was 44.

DOD -13 (in the case when the octyl group is a 2- ethyl hexyl group)

The surface tension (0.25 % solution, 20 °C stacto method) was 27.12 dyne / cm

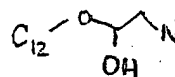
The phenol coefficient of a 10 % solution for typhoid bacteria was 42.

Actual Example 2

206 parts of di- ethylene tri amine was heated to 90 °C, and 139 parts of 2-dodecyl -2'-

chloro hydrin ether was gradually added into this, and it was reacted for 4 hours at 100 to 110 °C. After the reaction was completed, the contents were transferred to a separation funnel, and the amine hydrochloric acid salt that was by-produced was removed, and the oil-like material in the upper layer was distilled under reduced pressure, and thus, – mono (3- dodeca oxy -2- hydroxy propyl) di- ethylene tri amine was obtained.

Amine value 465.0, Molecular weight of 362



9.5 parts of mono chloro acetic acid was dissolved in 40 parts of water, and this solution was gradually added into 36.2 parts of this intermediate material, and it was reacted for 2 hours at 80 to 90 °C, and thus, the – mono (3- dodeca oxy -2- hydroxy propyl) di- ethylene tri amino acetic acid □ hydrochloric acid salt (from now on, this will be called MLD - 11) was obtained.

The surface tension (0.25 % solution, 20 °C stacto method) was 24.78 dyne / cm

Foaming strength (0.25 % solution, 20 °C ^{Ross miles?} ~~Lesmiles?~~ method) 245 mm

The phenol coefficient for typhoid bacteria was 89.

Actual Example 3

19.3 parts of di- ethylene tri amine was heated to 160 °C, and 35 parts of 3- (3- octa oxy -2- hydroxy propa oxy) -2- hydroxy propyl chloride was gradually added, and thereafter it was reacted at 170 to 180 °C for 3 hours. After the reaction was completed, it was extracted with ether, and the intermediate material which was mainly N, N' bis [3- (3- octa oxy -2- hydroxy propa oxy) -2- hydroxy propyl] di- ethylene tri amine was obtained.

Amine value 362.5, Molecular weight of 464

8.1 parts of mono chloro acetic acid was dissolved in 50 parts of iso propanol, and this

solution was gradually added into 40 parts of this intermediate material, and it was reacted for 2 hours at 80 °C, and thus, the N, N' bis [3- (3- octa oxy -2- hydroxy propa oxy) -2-hydroxy propyl] di- ethylene tri amino acetic acid □ hydrochloric acid salt (from now on, this will be called DOD - 21) was obtained.

The surface tension (0.25 % solution, 20 °C stacto method) was 27.21 dyne / cm

Foaming strength (0.25 % solution, 20 °C Losmiles? method) 167 mm

The phenol coefficient for typhoid bacteria was 110.

Actual Example 4

40 parts of 3- octa thio -2- hydroxy propyl chloride was reacted with 25.9 parts of di- ethylene tri amine the same as in Actual Example 1, and the intermediate material which was mainly N, N' bis (3- octa thio -2- hydroxy propyl) - di- ethylene tri amine was obtained. Mono chloro acetic acid was reacted with this intermediate material, and the N, N' bis (3- octa thio -2- hydroxy propyl) - di- ethylene tri amino acetic acid □ hydrochloric acid salt was obtained.

The phenol coefficient for typhoid bacteria was 170.

Actual Example 5

120 parts of 3- octa oxy -1, 2- di- chloro propane was reacted with 77 parts of di- ethylene tri amine the same as in Actual Example 1, and the obtained intermediate material was reacted with mono chloro acetic acid, and the N, N' bis (3- octa oxy -2- chloro propyl) - di- ethylene tri amino acetic acid □ hydrochloric acid salt was obtained.

The phenol coefficient for typhoid bacteria was 330.

Actual Example 6

111 parts of 2-octyl-2'-chloro hydrin ether was reacted with 142 parts of tetra ethylene penta amine the same as in Actual Example 1, and the intermediate material which is mainly N, N' bis (3-octa oxy -2- hydroxy propyl) - tetra ethylene penta amine was obtained. Mono chloro acetic acid was reacted with this intermediate material, and N, N' bis (3-octa oxy -2- hydroxy propyl) - tetra ethylene penta amino acetic acid \square hydrochloric acid salt (from now on, this will be called DOT - 11) was obtained.

The surface tension (0.25 %, 20 °C stacto method) was 26.97 dyne / cm

Foaming power (0.25 %, 20 °C Losmiles? method) 168 mm

The phenol coefficient for typhoid bacteria was 33.

Actual Example 7

28. 8 parts of di-ethylene tri amine was heated to 160 °C, and 70 parts of octyl glycidyl ether was slowly added, and it was reacted at 180 °C for 2 hours, and the intermediate material which was mainly N - mono- (3-octa oxy -2- hydroxy propyl) di-ethylene tri amine was obtained.

Amino value 537, Molecular weight of 313

16.2 parts of mono chloro acetic acid was dissolved in 66 parts of water, and this solution was gradually added into 50 parts of this intermediate material, and it was reacted for 3 hours at 90 to 100 °C, and thus, the N - mono- (3-octa oxy -2- hydroxy propyl) di-ethylene tri amino acetic acid \square hydrochloric acid salt (from now on, this will be called MOD - 11) was obtained.

MOD - 11 (In the case when the octyl group was a 2-ethyl hexyl group)

The phenol coefficient of a 10 % solution for typhoid bacteria was 39.

The results of the skin irritation test for the disinfectant surfactant of this invention

obtained in the above mentioned manner, and those of the existing, commercially sold product are shown in table 2.

Table 2. Results of Skin Irritation Test of Disinfectant Surfactant

Composition of material tested		Concentration applied	Rabbit skin irritation index *
Compound	Content		
Benzalkonium chloride (J P VII)	10 % (commercially sold product)	10 X diluted solution	100
Mono lauryl di- ethylene tri amino acetic acid	30 % (commercially sold product)	30 X diluted solution	121
DOD - 11 (in the case when octyl group is - octyl group)	10 %	10 X diluted solution	59.4
DOD - 21	10 %	10 X diluted solution	1.3
MLD - 11	10 %	10 X diluted solution	71.8
DOT - 11	10 %	10 X diluted solution	45.5

*** Rabbit skin irritation test**

The hair of the rabbit was shaved from its back and stomach side at 7 to 8 places on each side, and the testing solution was applied on these spots twice a day for 7 days, and it was visually observed with the naked eye once a day for 10 days from 1 day after application, and it was evaluated. The test was done with 3 healthy rabbits (2.0 to 2.5 kg each) as one group.

Evaluation :

No change:	0
Applied position became slightly red:	1
Redness can be clearly recognized on the applied position:	2
Redness and thickness can be recognized in the applied position:	3
Necrosis can be recognized in the applied position:	4

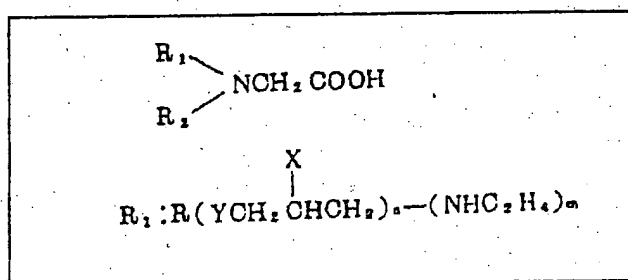
The value of each tested material was calculated by multiplying the number of days applied, F : 10, 9, 8,, 1, by this evaluation number.

Rabbit skin irritation index =

[(value of each tested material) / (value of 10 % Benzalkonium chloride solution)] x 100

Scope of the Patent Application

1) The disinfectant surfactant which is comprised of the compound that can be indicated by the below listed general formula, and an inorganic acid salt or an organic acid salt, or an alkali salt of these.



R₁ :

R₂ : a hydrogen atom or R₁

[Here, R is an alkyl group with 6 to 16 carbon atoms, or an alkyl allyl group. Y is an oxygen atom or a sulfur atom. X is a hydroxyl group or a halogen atom. n is an integer of 1 to 3, and m is an integer of 1 to 5.]

